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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "The importance of crystalline phases in ice nucleation by volcanic ash" *by* Elena C. Maters et al.

Anonymous Referee #1

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SUMMARY

In this work, Maters et al. conducted immersion-freezing ice nucleation experiments on 9 separate samples of volcanic ash. The ashes were chosen to span a range of chemical compositions, crystallinities, and mineralogies. Central to this paper, the authors adapted a technique from volcanology to probe the underlying cause of volcanic ash ice nucleation. In this technique, the ash samples are heated to 1400-1600 C to melt the mineral components of the ash samples. The heated samples were then allowed to cool to room temperature, where the hot ash quenched into a glass and impeded mineral nucleation and growth. The untreated and heated/quenched samples are defined as tephra and glass, respectively. The authors found that, despite being almost identical in chemical compositions, the tephra and glass had vastly different ice

nucleation abilities. Specifically, the tephra always nucleated ice more efficiently than the glass.

Overall, this paper is well written, the techniques used in the paper are well established, and the results are well supported. Furthermore, this work is well within the scope of ACP, and presents novel data to the field of ice nucleation. This reviewer, however, had several qualms with the interpretation of the results. As a general comment, the results from this manuscript represent an incredibly interesting and complex data set. There is a lot of thought-provoking behavior to unpackage, and the reviewer feels like the authors have not fully discussed the results–perhaps because they were beyond their original hypotheses. Instead, the reviewer feels like the authors have jumped to qualitative conclusions–many of which are poorly supported by their experiments. In addition, the authors do a poor job of putting their work in the context of previous works outside of their laboratory. Those comments are outlined below in the "Major Comments" section.

MAJOR COMMENTS

The title of the paper emphasizes the importance of crystalline phases. This, while qualitatively true, is poorly supported by the quantitative techniques in this paper. For example, the LIPteph, LIPglass, CIDteph, and CID glass are all devoid of crystalline material (<2%, i.e., below the limit-of-detection of the instrument), yet they all have vastly different ice nucleation abilities. In addition, their "glass" examples generally differ from their "teph" case. These are two of the nine cases, so they are over 20% of the samples. Ultimately, that some ice active mineral components (or at least 1400-1600 C labile components) that are present at less than 2% is an interesting result from this work that needs further highlighting.

Page, 5, Line 26: In this work alone, there is much evidence that Na/Ca-feldspar is not responsible for ice nucleation. For example, the plagioclase parameterizations from Harrison et al. (in prep) are much too low to explain the ice nucleation efficiency of almost all of the ash samples in this work, regardless of their plagioclase content. Furthermore, while Harrison et al., 2016 have shown that higher Na2O/CaO may imply higher ice nucleation activity for some feldspars, Page 5, Line 40 is direct contrast to previous studies that have shown that very pure albite is not that ice nucleation active [e.g., Zolles et al., 2015, Schill et al., 2015, Welti et al., 2019]. Alternatively, while there is less literature evidence that orthopyroxene may be the responsible agent, it seems like a much more feasible choice here. It is interesting to the reviewer that the authors then suggest in the conclusion that intermediate to felsic alkaline magmas may then have a higher propensity to contain ice-active ash in their eruptions.

Page 6, Line 1. The electron microprobe studies are not described in the text or in the supplemental. I see from Text S1 that the spot size is $\sim$10 um; however, it would be useful to know some quantitative limits on this technique as well as how many spot sizes per sample were looked at.

Page 6, Line 9. I am confused by this paragraph. The authors spend a great deal of time describing why LACteph does not have perthitic intergrowth microtexture, but then end the discussion by stating that NUOteph and ASTteph also don't have perthitic intergrowth microtexture. This seems like a logical fallacy to me. A similar sentiment is felt for the section on the anti-rapakivi texture. Was it not observed in the LACteph sample? Finally, how are all of these these surfaces susceptible to changes upon milling with a zirconia ball and vial?

MINOR/TECHNICAL COMMENTS

Page 1, Line 10: This abbreviation seems slightly confusing in the context of ice nucleation, since Vali et al. 2014 have proposed the acronym INE as "ice nucleating entity." I suggest you change INE to something like INeff"

Page 1, Line 15: "warmer" instead of higher?

Page 1, Line 20: The word "categorically" seems excessive here.

Page 1, Line 28: The sentence that starts "Ice formation" is unnecessarily long. I would suggest splitting into at least two sentences. A natural break in theme occurs at "as well as."

Page 1, Line 36: This sentence seems to be missing a comma and coordinating conjunction after diameter. It is the volcanic ash that is usually dominated, not the diameter that is usually dominated.

Page 1, Line 36: This paragraph seems incredibly weak. Part of the problem is that many statements are weakly lumped into "and references therein." For example, there is little mention of previous work on volcanic ash. For something like dust with hundreds of studies converging on a typical behavior, this could be appropriate–however; there are few previous experiments on volcanic ash, and each of them add holistically to the story presented here.

Page 2, Line 14: I would prefer that this sentence, if left here, explains how you "improv[ed] the understanding" instead of just simply stating that it will be done.

Page 2, Line 22: Since this is an atmospheric chemistry and physics journal, and not a geology journal, it would be helpful for readers of this journal to have the melting point range of each of the minerals in Table 2 compiled for them. That would greatly help them interpret the results of this study and perhaps elucidate why some samples retain some ice nucleation activity after the melt/quench cycle.

Page 3, Line 28 and Line 37: These equations need commas after them, since you have another clause after them (starting with "where").

Page 4, Line 10: I see why these arguments are included in this section, but they seem out of place. For example, the glassy organic particles all nucleated ice in the "deposition" mode and were certainly not immersed in water droplets at or above water saturation. I would suggest the first two sentences of this paragraph be qualified or removed.

Page 4, Line 16: As the figure looks now, there does not seem to be significant overlap with the markers and the grayed out region. Perhaps adding error bars to all curves would make that more clear? Or put error bars on the points in Figure 3?

Page 6, Line 28: Or, potentially, mafic magmas with high orthopyroxene?

Page 7, Line 20: There should be a comma between orthopyroxene and which.

Page 7, Line 37: But–isn't the ash you collected from volcanic plumes where high concentrations of acidic gases likely already interacted with the ash?

Figure 1 Legend: Is LEI in Figure 1 KIL everywhere else?

Figure 2, 3, and 5. The symbols in these figures are unreasonably small, especially in print form. This makes it relatively difficult to see the different between some of the samples that have similar marker colors (e.g., green, red, light blue.)

ADDITIONAL REFERENCES

Zolles T. et al., [2015], J. Phys. Chem. A, 119 (11), 2692-2700, DOI: 10.1021/jp509839x

Welti, A., Lohmann, U., and Kanji, Z.A., [2019], Atmos. Chem. Phys. Discuss., DOI: 10.5194/acp-2018-1271

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